AD-A156 025 NOISE IN SODIUM BETA ALUMINA CRYSTALS(U) UTAH UNIV SALT LAKE CITY DEPT OF PHYSICS J J BROPHY ET AL. SEP 85 TR-7 N00014-82-K-0603 1/1

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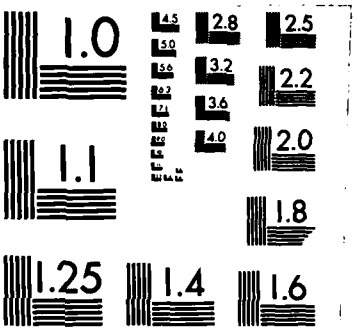

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

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1. REPORT NUMBER: Seven (7)	2. GOVT ACCESSION NO.: AD-A156 025
3. RECIPIENT'S CATALOG NUMBER: 5-28242	
TITLE (and Subtitle): Noise in Sodium β" Alumina Crystals	5. TYPE OF REPORT & PERIOD COVERED: Technical Report #7
	6. PERFORMING ORG. REPORT NUMBER: -
AUTHOR(s): James J. Brophy and Steven W. Smith	8. CONTRACT OR GRANT NUMBER(s): N00014-82-K-0603
PERFORMING ORGANIZATION NAME AND ADDRESS: Physics Department University of Utah, Salt Lake City, Utah 84112	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: -
CONTROLLING OFFICE NAME AND ADDRESS: ider, Chemistry Division, Associate Director of hematic & Physical Sciences, Office of Naval search, 800 N. Quincy St., Arlington, VA 22217	12. REPORT DATE: September 1985 13. NUMBER OF PAGES: ten (10)
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office): Office of Naval Research Resident Representative, University of California, 239 Campbell Hall, Berkeley, CA 94720	15. SECURITY CLASS. (of this report): Unclassified 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

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17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

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18. SUPPLEMENTARY NOTES

-

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Diffusion noise, conductivity fluctuations, superionic conductors, beta alumina.

1.5/f beta

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

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S/N 0102-LF-014-6601

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

OFFICE OF NAVAL RESEARCH

Cntract #N00014-82-K-0603

TECHNICAL REPORT No. 7

NOISE IN SODIUM β" ALUMINA SINGLE CRYSTALS

by

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Prepared for Presentation

at the

8th International Conference on Noise in Physical Systems
Rome, Italy

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September 1985

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Experimental current noise spectra for single crystal sodium β" alumina are essentially identical to those previously reported for polycrystalline ceramic specimens. The measured noise power is proportional to the square of the dc sample current and varies as $f^{-1.5}$, which suggests conductivity fluctuations arising from diffusion of the mobile ions. Only a small fraction of the mobile ions appear to participate in the diffusion noise process and the number is thermally activated with an activation energy of 0.8eV, in agreement with data from ceramic specimens.

1. INTRODUCTION

Conductivity fluctuations in polycrystalline ceramic sodium β" alumina have been observed and interpreted in terms of diffusion noise of the mobile sodium ions[1]. This interpretation implies, however, that the granular nature of the ceramic does not influence diffusion noise and that only a small fraction of the mobile ions participate. The present work examines electrical noise in single crystal sodium β" alumina under conditions essentially identical to those used to study ceramic specimens. In addition, preliminary data on noise of single crystal silver β" alumina are compared to sodium conductors.

2. EXPERIMENTAL TECHNIQUE

Single crystal sodium β" alumina samples grown by the flux evaporation method are cut to an approximately square shape, 1x1x0.2 cm^3. The corners are sealed into the sides of four plastic test tubes to provide diagonally opposing corner current electrodes and transverse noise terminals. Two mole percent amalgam electrodes prepared by electrodeposition from an 80% aqueous solution of NaCl into mercury are introduced into all four test tubes. This technique provides ohmic, low noise contacts and is essentially the same configuration previously employed to examine ceramic specimens[1]. Aqueous 5-M silver nitrate electrodes are used in the case of single crystal silver β" alumina samples[2].

Transverse noise voltages are measured with a PAR 113 preamplifier. The preamplifier output is analyzed digitally using an A/D converter and FFT routine developed for the Apple IIe personal computer[3]. The system accurately measures Nyquist noise of resistances from 10^4 to $2x10^8$ ohms over the frequency range 10^{-4} to 10^4 Hz. Current is supplied to the current terminals from a filtered battery source through a 10^5 ohm metallic resistor.

3. EXPERIMENTAL RESULTS

Representative transverse noise voltage spectra are shown in Figure 1. The zero-

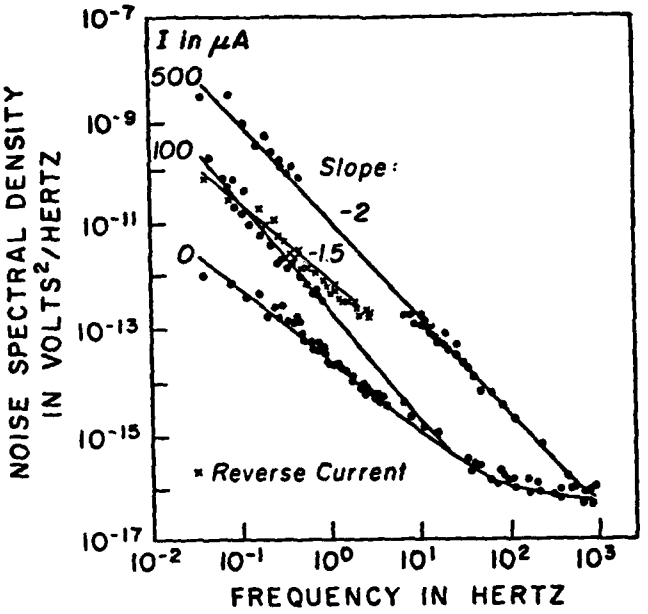

Figure 1. Transverse noise spectra of a sodium β" alumina single crystal.

current spectrum is attributed to chemical reaction noise at the crystal-amalgam interface similar to that seen in ceramic specimens[1,4]. The contact noise magnitudes and spectral shapes are the same for both types of samples and the single crystal contact noise decreases with time as previously reported for ceramics. At high

freqencies, the measured noise level agrees with Nyquist noise of the sample resistance, as determined from the slope of the (linear) current-voltage characteristic[4].

Current noise spectra are proportional to the square of the dc sample current and show spectral shapes ranging from $f^{-1.5}$ to $f^{-2.0}$, depending upon the duration of current in the sample. Lower values are seen accompanying the initial current levels and values of f^{-2} are noted after the extended current durations required to obtain full spectra. As in ceramic samples[1], the change is attributed to electrochemical changes accompanying current in the sample. Significantly, the spectral slope reverts to $f^{-1.5}$ upon current reversal, Figure 1.

Only one single crystal silver β'' alumina sample has been examined. The small size, 0.5x0.3x0.1 cm^3 precludes the use of the four-terminal method, but evidence from ceramic specimens[2] indicates that contact current noise is negligible, so that a two-terminal measurement may be acceptable. The observed noise is about the same as for ceramic samples and the spectral shape, $f^{-1.5}$, is very stable with both time and current.

The temperature dependence of the noise is different in different current and frequency regimes, Figure 2. At zero current

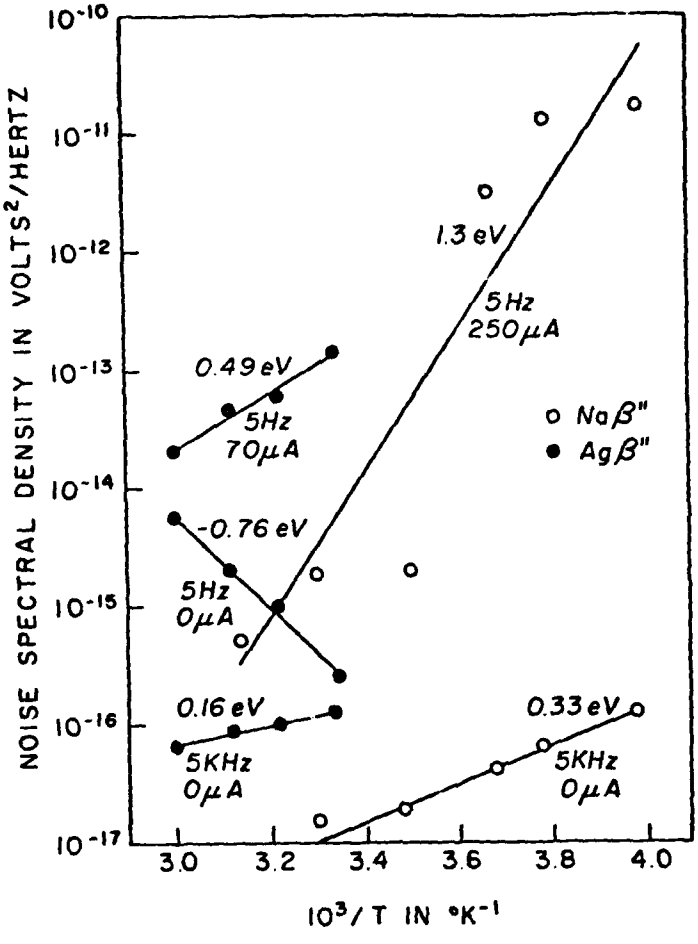

Figure 2. Temperature dependence of Nyquist noise (0μA, 5 kHz), current noise (70 or 250μA, 5 Hz), and contact noise (0μA, 5 Hz), for sodium and silver β" alumina single crystals.

and high frequencies the sodium β" alumina Nyquist noise is thermally activated with an activation energy of 0.33eV, in agreement with ceramic samples[1] and conductivity data[5]. Similarly, the silver β" alumina activation energy, 0.16eV, agrees with ceramic[2] and conductivity results[6].

The current noise is also thermally activated, with activation energies of 1.3eV and 0.49eV for sodium and silver β" alumina, respectively. The former value is in good agreement with ceramic sample data[1], while the corresponding figure for the silver ceramic is uncertain. Figure 2 also shows an activation energy of -0.76eV for contact noise in the case of silver β" alumina. The negative sign is consistent with a thermally-activated chemical reaction at the interface.

4. DISCUSSION

The observed spectral shape, $f^{\sim 1.5}$, is consistent with ceramic sample data, and with diffusion noise[7]. The noise voltage

spectral density for conductivity fluctuations arising from diffusion can be written as

$$\frac{S(V,f)}{V^2} = 4\,\frac{\langle \Delta N^2 \rangle}{N}\left(\frac{D}{2L^2}\right)^{1/2}\omega^{-3/2} \qquad (1)$$

Where V is the dc voltage across the sample, $\langle \Delta N^2 \rangle$ and N are the variance and average number of the diffusing ions, D is the diffusion constant, L is a characteristic length, and ω is the angular frequency. This expression is valid for frequencies greater than a characteristic frequency ω_0,

$$\omega_0 = 2D/L^2 \qquad (2)$$

Below ω_0, the spectrum flattens.

Taking D = 1×10^{-6} cm^2/sec at room temperature[6] and L equal to the sample length, 1 cm, the characteristic frequency is calculated to be 3.2×10^{-7} Hz, well below the range of present data. No characteristic frequency is observed for ceramic

specimens either[1], which indicates that the polycrystalline granularity is not important in determining the characteristic length, L.

Using experimental values of $S(V,f)/V^2$, and assuming Poisson statistics, $\langle \Delta N^2 \rangle = N$, a value for the mobile ion density of 1.2×10^8 ions/cm^3 is calculated from Equation 1. This is much smaller than the known concentration of mobile sodium ions, about 10^{22} ions/cm^3, but is in agreement with data from ceramic samples.

Following the approach taken in the case of the ceramic specimens[1], the temperature dependence of the noise can be accounted for by assuming

$$D(T) = D_0 \exp(-E_d/kT)$$
$$R(T) = R_0 T \exp(E_d/kT)$$
$$N(T) = N_0 \exp(-E_n/kT) \qquad (3)$$

so that Equation (1) becomes

$$S(V,f,T) = \frac{4I^2 R_0^2}{N_0} \left(\frac{D_0}{2L}\right)^{1/2} T^2 \omega^{-3/2} \times$$

$$\times \exp[(3E_d/2 + E_n)/kT]$$

Now, from Figure 2, $(3E_d/2 + E_n) = 1.3$eV

and E_d = 0.33eV, so that E_n = 0.8eV. Inserting this and the calculated room temperature value of the ion density into Equation 3, $n_o = 3.5 \times 10^{21}$ ions/cm^3. This is close to the expected number of mobile Na ions.

5. CONCLUSIONS

This experimental data indicates that conductivity fluctuations in single crystal sodium β" alumina and silver β" alumina are very similar to those observed in polycrystalline ceramic specimens. In particular, it appears that the granularity of ceramic specimens does not influence the diffusion noise, so that future studies may concentrate on ceramic samples, which are more plentiful than single crystals. The observed experimental noise level can be accounted for by assuming that the number of mobile ions participating in the diffusion noise process is thermally activated.

6. ACKNOWLEDGMENTS

This research is supported in part by the Office of Naval Research. The authors express their appreciation to G. R. Miller and J. M. Viner for many helpful suggestions and advice.

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